PB-246 311

EVALUATION OF POLLUTION CONTROL IN FOSSIL FUEL CONVERSION PROCESSES. COAL TREATMENT: SECTION 1. MEYERS PROCESS

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Linden, New Jersey

September 1975

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The report gives results of a review of the Meyers process being developed by TRW, Inc., from the standpoint of its potential for affecting the environment. The quantities of solid, liquid, and gaseous effluents have been estimated, where possible, as well as the thermal efficiency of the process. For the purpose of reduced environmental impact, a number of possible process modifications or alternatives which could facilitate pollution control or increase thermal efficiency have been proposed, and new technology needs have been pointed out.

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TABLE OF CONVERSION UNITS

To Convert From	To	Multiply By
Btu	Calories, kg	0.25198
Btu/pound	Calories, kg/kilogram	0.55552
Cubic feet/day	Cubic meters/day	0.028317
Feet	Meters	0.30480
Gallons/minute	Cubic meters/minute	0.0037854
Inches	Centimeters	2.5400
Pounds	Kilograms	0.45359
Pounds/Btu	Kilograms/calorie,kg	1.8001
Pounds/hour	Kilograms/hour	0.45359
Pounds/square inch	Kilograms/square centimeter	0.070307
Tons	Metric tons	0.90719
Tons/day	Metric tons/day	0.90719

1. SUMMARY

The Meyers process being developed by TRW, Inc. has been reviewed from the standpoint of its potential for affecting the environment. The quantities of solid, liquid and gaseous effluents have been estimated, where possible, as well as the thermal efficiency of the process. For the purpose of reduced environmental impact, a number of possible process modifications or alternatives, which could facilitate pollution control or increase thermal efficiency, have been proposed; and new technology needs have been pointed out.

2. INTRODUCTION

Along with improved control of air and water pollution, the country is faced with urgent needs for energy sources. To improve the energy situation, intensive efforts are under way to upgrade coal, the most plentiful domestic fuel, to liquid and gaseous fuels which give less pollution. Other processes are intended to convert liquid fuels to gas. A few of the coal gasification processes are already commercially proven, and several others are being developed in large pilot plants. These programs are extensive and will cost millions of dollars, but this is warranted by the projected high cost for commercial gasification plants and the wide application expected in order to meet national needs. Coal conversion is faced with potential pollution problems that are common to coal-burning electric utility power plants in addition to pollution problems peculiar to the conversion process. It is thus important to examine alternative conversion processes from the standpoint of pollution and thermal efficiencies and these should be compared with direct coal utilization when applicable. This type of examination is needed well before plans are initiated for commercial applications. Therefore, the Environmental Protection Agency arranged for such a study to be made by Exxon* Research and Engineering Company under contract EPA-68-02-0629, using all available non-proprietary information.

The present study under the contract involves preliminary design work to assure that conversion processes are free from pollution where pollution abatement techniques are available, to determine the overall efficiency of the processes and to point out areas where present technology and information are not available to assure that the processes are non-polluting.

All significant input streams to the processes must be defined, as well as all effluents and their compositions. This requires complete mass and energy balances to define all gas, liquid, and solid streams. With this information, facilities for control of pollution can be examined and modified as required to meet Environmental Protection Agency objectives. Thermal efficiency is also calculated, since it indicates the amount of waste heat that must be rejected to ambient air and water and is related to the total pollution caused by the production of a given quantity of clean fuel. Alternatively, it is a way of estimating the amount of raw fuel resources that are consumed in making the relatively pollution-free fuel. At this time of energy shortage this is an important consideration. Suggestions are included concerning technology gaps that exist for techniques to control pollution or conserve energy. Maximum use was made of the literature and information available from developers. Visits with some of the developers were made, when it appeared warranted, to develop and update published information. Not included in this study are such areas as cost, economics, operability, etc. Coal mining and general offsite facilities are not within the scope of this study.

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* Prior to June 1, 1974 Exxon Research and Engineering Company conducted business under the name Esso Research and Engineering Company.

Our previous studies in this program to examine environmental aspects of fossil fuel conversion processes covered various methods for coal conversion to clean fuels. Reports have been issued on both gasification and liquefaction processes including Koppers-Totzek, Synthane, Lurgi, CO_2 Acceptor, COED, SRC and BI-GAS processes (1,2,3,4,5,6,7). The present report extends these studies to include chemical cleaning of coal by the Meyers process being developed by TRW, Inc. under contract to EPA. In this process pyritic sulfur is removed from coal by the action of a solution of ferric sulfate. The coal is not "converted," and it essentially retains its original heating value. The pyritic sulfur leaves the process as elemental sulfur and iron sulfates.

We wish to acknowledge the information and assistance provided by the Environmental Protection Agency. This study is based, to a large extent, on information supplied by EPA and on reports published by TRW, Inc. for EPA.

3. PROCESS DESCRIPTION

In the Meyers process, the pyrites in the coal are removed by reaction with ferric sulfate in a solution containing ferric and ferrous sulfates and sulfuric acid. The ferric ion is continuously regenerated by reaction of oxygen and ferrous ion. The elemental sulfur product is extracted with an organic solvent. The iron product from the pyrites is removed as solid ferric and ferrous sulfates.

A block flow diagram of the basic Meyers process is shown in Figure 1. The contents of the indicated streams are shown in Table 1. This diagram and description are based on information supplied by EPA (in the form of a process flow sheet produced by Dow Chemical U.S.A.) and EPA reports (8,9). Later evaluations of the process may be available but for the present study no improvement would be expected in the results without pilot unit data. The process description is not complete without utilities, coal preparation and storage, etc. These items will be addressed later in this report.

A more recent design for a commercial plant has been completed by Dow Chemical U.S.A.[12] That design has a more conservative approach to heat integration than the one used in the present work, produces a drier, compacted product and shows electricity to be purchased.

3.1 Reactor Section

Coal that has been ground to less than 100 mesh (Str. 2) is mixed with recycled leach solution (Strs. 8, 11, 13) in a flow through mixing tank. The mixing vessel is maintained at about 210°F. The slurry is continually pumped from the mixing vessel to one of 10 reactor vessels.

In the reactor vessels, the slurry is contacted with oxygen at about 300°F. The pyritic sulfur is 95% converted to elemental sulfur and sulfate in the reactor vessels. The reactions taking place in the reactors are shown below:

Leaching Reactions

(1) $FeS_2 + Fe_2(SO_4)_3 \longrightarrow 3FeSO_4 + 2S$

(2) $FeS_2 + 7Fe_2(SO_4)_3 + 8H_2O \longrightarrow 15FeSO_4 + 8H_2SO_4$

Since the net SO_4:S production from FeS_2 is approximately 1.5:1, the overall leaching reaction is:

(3) $FeS_2 + 4.6\ Fe_2(SO_4)_3 + 4.8\ H_2O \longrightarrow 10.2\ FeSO_4 + 4.8\ H_2SO_4 + 0.8\ S$

Regeneration Reaction

(4) $9.6\ FeSO_4 + 4.8\ H_2SO_4 + 2.4\ O_2 \longrightarrow 4.8\ Fe_2(SO_4)_3 + 4.8\ H_2O$

Net Overall Reaction

(5) $FeS_2 + 2.4\ O_2 \longrightarrow 0.2\ Fe_2(SO_4)_3 + 0.6\ FeSO_4 + 0.8\ S$

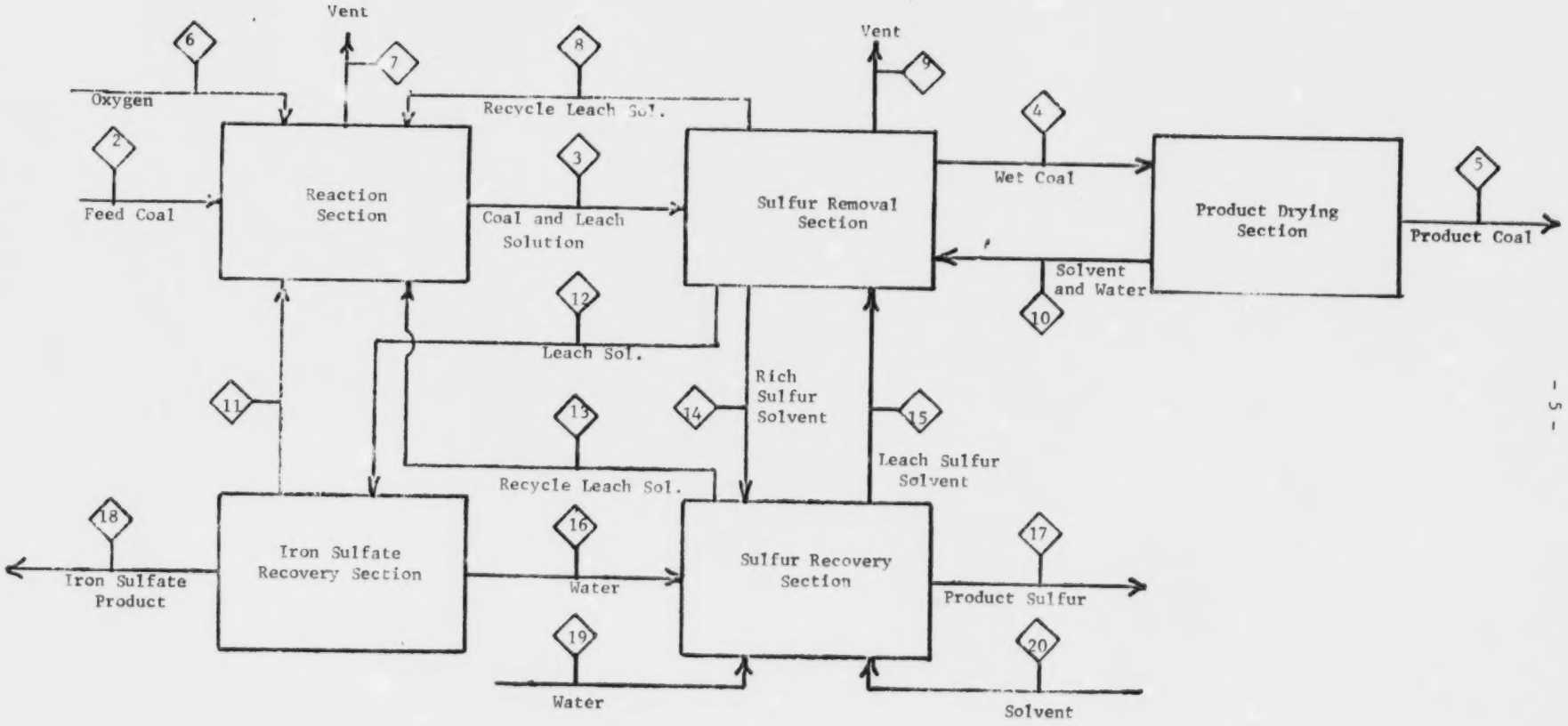

Figure 1

Original Basic Design of Meyers Process

Table 1

Stream Identification, Original Design, lb/hr

Stream	2	3	4	5	6	7	8	9	10	11
Coal*	188,000	188,000	187,800	187,800						
FeS_2	12,000	600	600	600						
S		2,400								
$FeSO_4$		30,000					18,000			1,000
$Fe_2(SO_4)_3$		122,800	200	200			73,800			31,600
H_2SO_4		6,800					4,000			2,200
H_2O	20,000	662,800	47,000	37,600		660	466,400	200	9,400	126,600
Solvent			9,400					200	9,400	
O_2					7,296					
Inert					40	40				
Total	220,000	1,013,400	245,000	226,200	7,336	700	562,200	400	18,800	161,400

* Feed coal ex pyrites and moisture.

Table 1 (Cont'd)

Stream Identification, Original Design, lb/hr

Stream	12	13	14	15	16	17	18	19	20
Coal	200						200		
FeS_2									
S			2,400			2,400			
$FeSO_4$	9,600	2,400	2,400				8,600		
$Fe_2(SO_4)_3$	39,600	9,800	9,800				7,400		
H_2SO_4	2,200	600							
H_2O	231,000	50,400	65,400	137,800	104,400			8,400	
Solvent			255,800	256,000					200
O_2									
Inert									
Total	282,000	63,200	336,400	393,800	104,400	2,400	16,200	8,400	200

The excess ferric and ferrous sulfates must be removed from the system. The slurry is cooled by heat exchange with fresh feed and then by cooling water and is pumped to the Sulfur Removal Section (Str. 3).

3.2 Sulfur Removal Section

In the Sulfur Removal Section, approximately 60% of the leach solution is removed in hydroclones and recycled to the Reaction Section (Str. 8). The remaining leach solution is removed by filtration and is passed to the Iron Sulfate Recovery Section (Str. 12).

The wet filter cake is washed with water and then mixed with recycle solvent (e.g., light naphtha) at 160°F and most of the elemental sulfur is dissolved. The resulting slurry is filtered to remove the cleaned coal which passes to the Product Drying Section (Str. 4). The sulfur-rich solvent is separated from water by decantation and passes to the sulfur recovery section (Str. 14).

3.3 Product Drying Section

The treated coal, containing about 25% moisture and 5% solvent (dry basis), is conducted to the drying section (Str. 4). The coal is partially dried under vacuum; the sensible heat of the coal is sufficient to remove all the solvent and about 20% of the water. The vapors are returned to the Sulfur Removal Section (Str. 10) where they are condensed in a water cooled vessel. The water and solvent are separated by decantation and reused in the process. The coal product, containing 20% moisture (dry basis) then leaves the process (Str. 5).

3.4 Sulfur Recovery Section

The sulfur-laden solvent and miscellaneous solvent and water streams are passed to the Sulfur Recovery Section (Str. 14). The solvent is removed from the sulfur by distillation and the sulfur leaves the process (Str. 17). Water and rich solvent are separated by decantation. The water is recycled to the Reaction Section (Str. 13) and the solvent is returned to the Sulfur Removal Section (Str. 15). Makeup water and solvent (Strs. 19 and 20) are added to the system through the Sulfur Recovery Section.

3.5 Iron Sulfate Recovery Section

The water filtrate from filtration in the Sulfur Removal Section passes to the Iron Sulfate Recovery Section (Str. 12). Since the process produces iron from the pyrites, it is necessary to remove iron from the system. The filtrate is heated to about 265°F, and some of the water is flashed overhead. Part of the steam thus formed is returned to the Reaction Section (in Str. 11) and part passes to the Sulfur Recovery Section in stream 16. The remaining slurry of iron sulfates is filtered at 215°F to produce an iron sulfate filter cake for disposal. The filtrate is returned to the Reaction Section.

4. MODIFIED PROCESS DESIGN INCLUDING UTILITIES AND EFFLUENTS

To more carefully assess the pollution potential and the thermal efficiency of the Meyers Process, an oxygen plant, coal preparation and storage and utilities have been included in the design and other slight changes have been made. The complete block diagram for the plant is shown in Figure 2 and the streams are identified in Table 2. An asterisk (*) indicates streams released to the environment.

4.1 Major Design Modifications

The plant has been made self sufficient with regard to all utilities. Steam and power are generated internally, water treatment facilities have been included, a cooling tower has been added to allow recycling the cooling water, the coal storage and preparation section was added and an oxygen plant has been assumed.

The coal analysis assumed in the present study is given in Table 3. This analysis corresponds to that of the Lower Kittanning coal given on page 10 of reference 8 except the pyritic sulfur content has been assumed to be 3.21% (dry basis) instead of 3.58% and the moisture has been assumed to be 10% to conform to previous design studies of the process (e.g., stream 1, Table 29, p. 120 of Ref. 8). Reference 9, p. 126, gives a pyritic sulfur content of 3.09%. Such changes will have little effect on the conclusions of this study.

4.2 Effluents to Air

4.2.1 Coal Storage and Preparation

ROM coal, 8 in. X 0, is received at the plant and stored. Three days storage (7920 tons, wet) has been suggested. This quantity of coal would probably be stored in silos with nitrogen blanketing. It would probably be advisable to store more coal (e.g., 30 days supply) in a "permanent" pile for emergency use. This pile could be covered with asphalt and used only in case of mine outage.

The ROM coal is conveyed to pulverizers where the coal is reduced to 80% less than 200 mesh. (This size is smaller than previous designs and is used to enhance reaction rates as well as to provide a product size suitable for combustion). The coal from the pulverizers is then fed to the Reaction Section.

It is not necessary to dry the coal as it is subsequently slurried in a water solution. It is assumed that covered conveyers will be used throughout to minimize dust problems. The coal dimunition equipment can be enclosed, with air vented to bag filters. This will reduce outside noise as well as provide for dust containment.

4.2.2 Reaction Section

Except for minor vents, which will be discussed later, no air effluents are emitted from the reaction section.

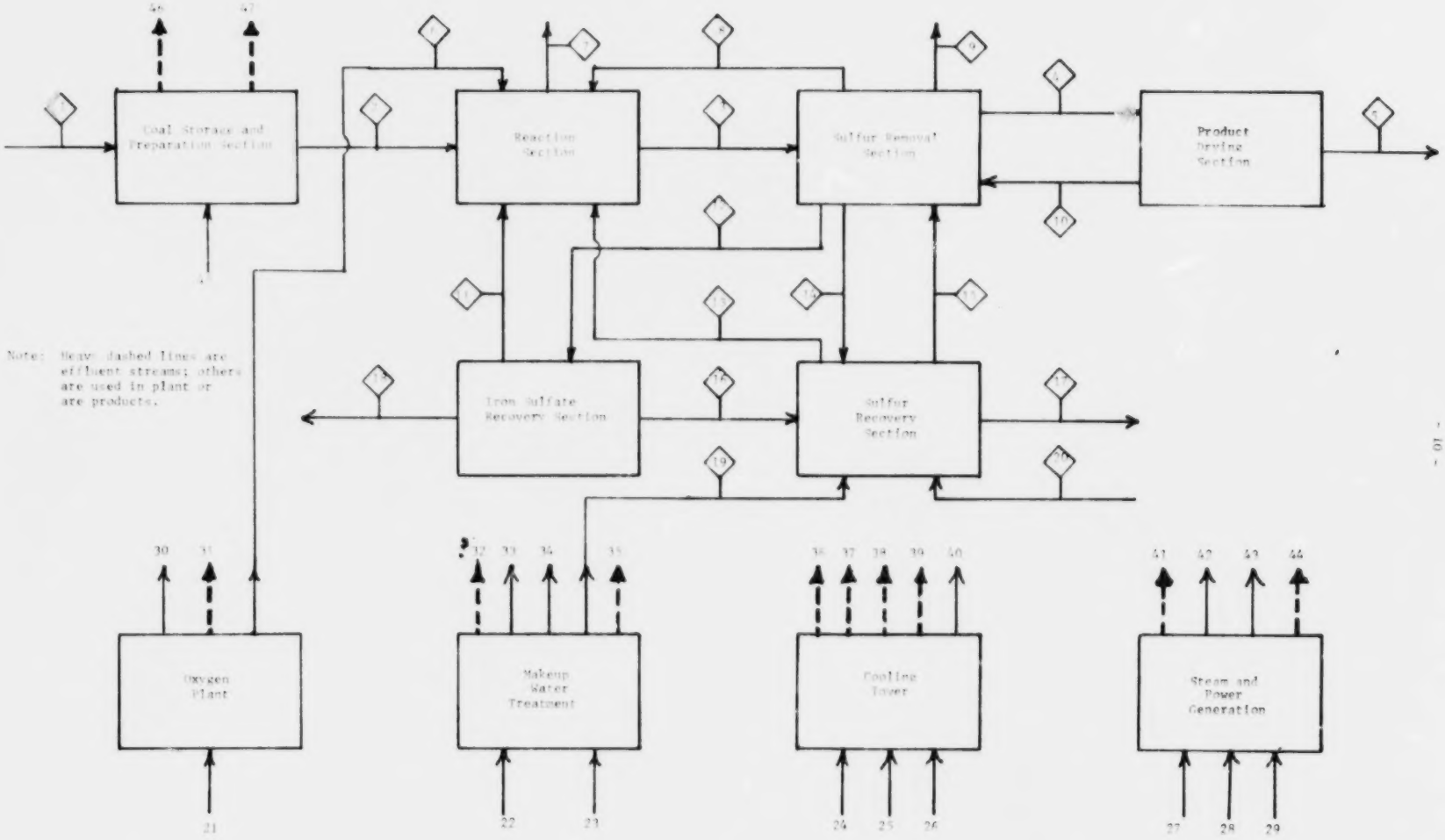
Coal Storage and Preparation Section
Reaction Section
Sulfur Removal Section
Product Drying Section
Iron Sulfate Recovery Section
Sulfur Recovery Section
Oxygen Plant
Makeup Water Treatment
Cooling Tower
Steam and Power Generation
Note: Heavy dashed lines are effluent streams; others are used in plant or are products.

Figure 2

Flow Diagram - Modified Meyers Process

Table 2

Stream Identification for Modified Process, Lb/Hr

Stream	1	2	3	4	5	6	7	8	9	10
Coal*	188,000	188,000	188,000	187,800	187,800					
FeS_2	12,000	12,000	602	602	602					
S			2,438							
$FeSO_4$			30,060					18,000		
$Fe_2(SO_4)_3$			122,798	200	200			73,800		
H_2SO_4			6,800					4,000		
H_2O	20,000	20,000	662,800	47,000	37,600		660	466,400	200	9,400
Solvent				9,400					200	9,400
O_2						7,296				
Inert						40	40			
TOTAL	220,000	220,000	1,013,498	245,020	226,220	7,336	700	562,200	400	18,800

* Feed coal, ex pyrites and moisture.

Table 2 (Continued)

Stream Identification for Modified Process, Lb/Hr

Stream	11	12	13	14	15	16	17	18	19	20
Coal		200						200		
FeS_2										
S				2,438			2,438			
$FeSO_4$	1,000	9,660	2,400	2,400				8,660		
$Fe_2(SO_4)_3$	31,600	38,998	9,800	9,800				7,398		
H_2SO_4	2,200	2,200	600	600						
H_2O	126,600	231,000	50,460	65,400	137,800	104,400			18,460	
Solvent				255,800	256,000					200
O_2										
Inert										
TOTAL	161,400	282,058	63,260	336,438	393,800	104,400	2,438	16,258	18,460	200

Table 2 (Continued)

Stream Identification for Modified Process, Lb/Hr

21 - Air to O_2 Plant	31,520	
22 - Chemicals	--	
23 - Water	153,850	
24 - Air	12,700,000 (4×10^9 scfd)	Air to Cooling Tower
25 - Water	135,560	Makeup Water to Cooling Tower at 85°F
26 - Water	7,083,600	Water plus makeup recirc. to Cooling Tower at 105°
27 - Product Coal	13,234 (Dry)	Product Coal to Utility Boiler
28 - Air	141,229	Air to Boiler
29 - Water	126,000	Boiler Feed Water (Includes 6000 lb/hr makeup water)
30 - Water	170	Moisture from Air to Boiler Feed Water
*31 - Nitrogen	24,050	Vent from O_2 Plant, can be used in coal silos
*32 - Sludge	--	From Treating Makeup Water
33 - Boiler Feed Water Makeup	5,830	To Steam Generation
34 - Cooling Water	129,560	Makeup
*35 - Backwash	--	
*36 - Air From Cooling Tower	12,700,000 (4×10^9 scfd)	Air from Cooling Tower
*37 - Drift Loss Water	14,160	Water Mist to Air
*38 - Water Vapor	100,000	Water Evaporated from Cooling Tower

Table 2 (Continued)

Stream Identification for Modified Process, Lb/Hr

*39 - Blowdown Water	21,400	Purge from Cooling Tower to Holding Pond
40 - Cooling Water	7,083,600	---
*41 - Flue Gas	154,570	From Utility Boiler
42 - Blow Down Water	6,000	To Cooling Tower Makeup
43 - Steam	120,000	
*44 - Ash	2,541	
45 - Rain**	e.g. 6" Rain in 24 Hours	Rain on Coal Storage and Preparation Area
*46 - Rain Run Off**	e.g. 6" Rain in 24 Hours	From Coal Storage and Preparation Area
*47 - Dust	--	From Coal Preparation Collect in Bag Filters

* These streams are emitted to the environment.

** Not applicable if all storage is in silos.

Table 3

Feed Coal Analysis (Dry Basis)

Proximate Analysis, Wt. %	
Fixed carbon	58.48
Volatile matter	20.66
Ash	20.86

Moisture = 10% wt. dry coal (assumed)

Heating value
HHV (dry) - 12140 Btu/lb

Ultimate Analysis, Wt. %	
Carbon	68.53
Hydrogen	3.85
Nitrogen	1.20
Chlorine	0.08
Sulfur	3.92
Ash	20.86
Oxygen (difference)	1.56

Sulfur Forms	%
Pyritic	3.21
Sulfate	0.04
Organic	0.67

4.2.3 Sulfur Removal Section

The only effluents in the Sulfur Removal Section come from vents which are discussed later. Pressure filtration is used, thus vapors are enclosed.

4.2.4 Product Drying Section

The naphtha solvent and a portion of the water are removed from the product coal by vacuum. The resulting vapors are condensed and returned to the process system.

The dry product coal heating value has been increased by ca 5% (Ref. 8), from 12,140 Btu/lb to 12,747 Btu/lb due to ash removal. With added sulfate and elemental sulfur the product contains approximately 0.95% sulfur in the following forms:

Sulfur Form	%
Pyritic	0.17
Elemental	0.04
Sulfate	0.03
Organic	0.71
	0.95%

(This is the analysis used in Ref. 8 but corrected for the decrease in coal weight in processing.)

No information is available on techniques available for drying the product coal. It is assumed that the product coal will be burned as is. Part of the coal (13,234 lb/hr, dry basis) is used in the steam plant. Thus, net product is 175,366 lb/hr (dry).

4.2.5 Iron Sulfate Recovery Section

Except for minor vents, there are no air effluents from the iron sulfate section. The filter is assumed to be enclosed to prevent vapors from escaping. It is also assumed that the product iron sulfates will be handled in a moist form to avoid dusting.

4.2.6 Sulfur Recovery Section

Again, this section is completely enclosed except for vents and there are no effluents to the air. If the sulfur product is stored as a liquid, there will be no emissions since the storage and handling facilities will be enclosed. If the product is handled and stored as a solid, then control of dust is necessary.

4.2.7 Auxiliary Facilities

The auxiliary facilities in the complex include an oxygen plant, raw water treatment, cooling towers and steam and power generating facilities. These auxiliary units must be considered to evaluate effluent problems and overall thermal efficiency.

The oxygen plant is a major consumer of power and there is a large gaseous effluent. It has been assumed in the present design that an extraction turbine, using 600 psig steam, is used to drive the air compressor in the oxygen plant. The extraction steam, at 115 psig, is utilized in the rest of the plant. The effluent to the air consists of 24,050 lb/hr (5435 cu ft/min) of relatively pure nitrogen which requires no cleaning.

Moisture containing air from the cooling tower represents the largest effluent to the atmosphere. In this particular plant the cooling water should be relatively free of volatile materials; pressures on the heat exchangers are low and, except for the organic solvent, no volatile organics have been reported as being present in the reaction system. Fog formation can sometimes represent a problem with cooling towers. The extent of this problem is determined in large part by the plant location. Drift loss from the cooling tower can cause dust problems when the solids in the cooling water are deposited. It is expected that cooling tower blowdown will be sent to an evaporation pond. Due to the nature of the present process, there should be no noxious fumes from this pond if there are no leaks in the naphtha heat exchangers.

A raw water treatment system is provided to furnish makeup water to the steam boiler and cooling tower. No air effluents are expected from this unit.

Product coal is burned in the steam plant and the flue gas represents the largest quantity of noxious contaminants emitted to the atmosphere. The combustion gas contains dust, NO_x, CO and sulfur compounds and these must be controlled. The use of product in the boiler furnace also affects the thermal efficiency of the overall plant. Control of particulate matter can be effected by the use of commercial electrostatic precipitators, cyclones and/or scrubbers. The use of excess air should reduce the CO content of the stack gas. NO_x emissions can be reduced by careful control of combustion conditions and staged firing. The ultimate limit on NO_x reductions has not been reached as considerable work is in progress on techniques for NO_x control. It is expected that a technique will be developed eventually for direct removal of NO_x from stack gases or for its conversion to N_2. This problem, however, could exist for a long time.

Sulfur in the flue gas represents a problem. The sulfur in the product coal produces 1.49 lb of SO_2 per million Btu. This is higher than the present Federal limit of 1.2 lb SO_2/MM Btu for large power plants. The major part of this sulfur in the fuel is in the form of organic sulfur (0.71% out of 0.95%) which is not removed in the process. In many coals, the organic sulfur content (and total sulfur in the product coal) is much higher than that shown here (Ref. 9, Appendix D). It is thus expected that,

for most coal feeds, the total sulfur in the product coal will be higher than that used here. Though the total sulfur has been decreased remarkably, the content would indicate the need for stack gas scrubbing on the boiler furnace stack. This, however, would defeat the purpose of using product coal for boiler fuel.

For some coals, the product may be used as fuel so that emissions meet Federal regulations for new installations. It has been estimated (10) that the percentage of Appalachian coals meeting the requirement of 1.2 lb SO_2 emissions per million Btu could be increased from about 10% to 40%. Due to variations in pyrites removal and increase in sulfate content, these coals would, of course, have to be tested. That some coals can be desulfurized to meet existing regulations has been recently reported (11).

4.2.8 Minor Vents

It is expected that the effluents from minor vents in the process will be collected. The moisture and solvent vapor will be condensed and returned to the system. The net vent gas can be incinerated in the utility furnace.

4.3 Liquid and Solid Effluents

4.3.1 Coal Storage and Preparation

The major liquid and solid effluents from this area consist of rain runoff and wash water from coal dust removal. This water should be sent to a storm pond where solids can settle out. If there are no spills of organic materials in the process area, storm drainage from this area can also be sent to this holding pond. After sufficient settling time, the water from this pond can be used as raw process water. The pH of the pond water can be corrected for acid content by limestone addition to the pond circuit or the treatment can be accomplished in the makeup Water Treatment Section. The pond should be large enough to prevent contamination by overflow and, if the soil is such that seepage would be a problem, the bottom of the pond should be lined with clay, concrete or other impervious material.

4.3.2 Reaction and Sulfur Removal Sections

There are no liquid or solid plant effluent streams from the Reaction and Sulfur Removal Sections.

4.3.3 Product Drying Section

The only effluent stream from the Product Drying Section is the product itself. This product contains part of the sulfur input to the plant, much of the minor element input and the major part of the trace element input. The water and solvent removed from the coal in this section are returned to the process. The further transportation, use or storage of the product is not specified, but enclosed transport should be used to prevent dusting problems.

4.3.4 Iron Sulfate Recovery Section

The effluent from the Iron Sulfate Recovery Section consists mainly of ferrous and ferric sulfate. The moisture content of this material, as well as its disposition, are unspecified. There doesn't seem to be a simple solution to the disposition problem as the material is water soluble and will be acidic. This problem is considered further under Potential Improvements.

4.3.5 Sulfur Recovery Section

The sulfur product is the only liquid or solid effluent from the Sulfur Recovery Section. In the present design, the sulfur leaves as a liquid. If the material is subsequently cooled to form a solid, care will have to be taken to prevent the escape of dust.

4.3.6 Auxiliary Facilities

The oxygen plant has no solid effluent and the only liquid effluent is water, condensed from the air, that is used for boiler feed makeup.

The effluent from the Makeup Water Treatment area consists of sludge and salt-laden water. These streams arise from the materials used for water treatment; lime, aluminum sulfate, caustic soda, sulfuric acid, etc. It is anticipated that the simplest method for disposal of these materials will be transport to the evaporation pond used for cooling tower blowdown. This pond is expected to be impervious to water and can handle the effluent from the water treating area.

Blowdown from the cooling tower is sent to this evaporating pond as previously indicated. This water should contain only non-volatile inorganic materials.

The liquid effluent from the Steam and Power Generation Section is boiler blowdown which is used for cooling tower makeup. The solid effluent consists of 2,541 lb/hr of ash which consists of the minor and trace elements in the original coal. This ash could be slurried with part of the blowdown water from the cooling tower and pumped to a mine for disposal. The ash could be disposed of in the pond used for cooling tower blowdown but the quantity is such that the pond size might be excessive.

5. THERMAL EFFICIENCY

The basic thermal efficiency of the Meyers' Process, using the present design, is estimated to be 92.1% (see Table 4). This is the heating value of the net product (total product less 13,234 lb/hr (dry) that is burned to generate steam) divided by the heating value of input coal. If the heating value of the by-product sulfur is included, the thermal efficiency rises to 92.5%. Results are summarized in Table 4.

It will be necessary to remove dissolved solids (other than iron sulfates) from the reaction section water circuit. One way of accomplishing this is to remove the iron sulfates in a water purge stream instead of filtering. It is then unnecessary to evaporate a large amount of water and the total thermal efficiency rises to about 92.6%. Debits (mainly, sulfuric acid makeup) occur, however, in this type of iron sulfate removal; these are discussed in Section 8.

Table 4

Thermal Efficiency

Net thermal efficiency	92.1%
Including sulfur product	92.5%
Without water evaporation in iron sulfate removal*	92.6%

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* See Section 8 for a description of this alternative and the debits thereby incurred.

6. SULFUR BALANCE

A detailed sulfur balance for the Meyers' process is shown in Table 5. In the present design, most of the sulfur exits the plant in solids; the boiler stack is the only source of gaseous sulfur emission. As indicated earlier, most of the sulfur in the coal product is in the form of organic sulfur not removed in the processing.

Table 5

Sulfur Balance

	lb/hr		Percent
Sulfur Into Plant			
In coal feed	7,836		100.0
Pyritic 6416		81.9	
Sulfate 80		1.0	
Organic 1340		17.1	
Sulfur Out of Plant			
In product coal	1,788		22.82
Net product coal 1,663		21.22	
Boiler stack gas 125		1.60	
In coal loss	2		0.03
In $FeSO_4$	1,828		23.33
In $Fe_2(SO_4)_3$	1,780		22.72
Product sulfur	2,438		31.11
Total sulfur from plant	7,836		100.0

7. TRACE ELEMENTS

The Meyers process offers an excellent example of how potentially hazardous trace elements can buildup in a coal treating plant. Reference to Figure 2 shows that the only egress of water from the process (other than minor vents) occurs with the product coal in Stream 5. (An unknown amount of water leaves with the iron sulfate in stream 18 as surface moisture and water of hydration.) The concentration of trace elements that are soluble will build up in the water recycled to the Reaction Section. Although the concentration can be limited by withdrawing a purge stream from the circuit, as indicated in a later section, the concentration level of dissolved solids is still somewhat arbitrary.

Analyses furnished by EPA for minor and trace elements in a Lower Kittanning coal are shown in Table 6 for both treated and untreated coal. The table indicates significant reductions in calcium, copper, iron, magnesium, manganese, nickel, potassium, sodium, sulfur, titanium and zinc. Some elements of interest, such as arsenic, are below the limits of analysis and whether or not these are dissolved is unknown.

Table 7 shows analyses for trace elements in a treated and untreated Lower Kittanning coal (reference 8, p. 189). Elements that are depleted by action of the pyrites leach solution are arsenic, boron, beryllium, chromium, copper, fluorine, manganese, nickel, selenium and zinc.

From the viewpoint that the product coal contains less of some potentially hazardous elements, leaching of these elements is beneficial. It should be kept in mind, however, that this leaching will concentrate these elements in a small area (depending on the method used for disposing of the purge from the reaction water circuit). More work is necessary to define potential problems from trace elements. A recycle system would be advantageous in ascertaining the extent of dissolved solids buildup.

Table 6

Minor and Trace Elements In
Coal Feed (Lower Kittanning)
and Product Coal (from EPA), ppm

Element	Method	Untreated	Treated
Aluminum	ES	19000.	22000.
Antimony	ES	<2.	<2.
Arsenic	ES	<2.	<2.
Beryllium	ES	<1.	<1.
Bismuth	ES	<1.	<1.
Boron	ES	66.	66.
Cadmium	AA	2.4	2.0
Cadmium	ES	<5.	<5.
Calcium	ES	1100.	440.
Chloride	NA	8600.	8500.
Chromium	AA	30.	32.
Chromium	ES	44.	42.
Cobalt	ES	11.	15.
Copper	AA	12.	7.0
Copper	ES	40.	12.
Fluoride	IE	17.	18.
Germanium	ES	22.	22.
Iron	AA	40000.	16700.
Iron	ES	33000.	19000.
Lead	AA	21.	29.
Lead	ES	80.	82.
Lithium	ES	9.	9.
Magnesium	ES	1300.	880.
Manganese	AA	28.	15.
Manganese	ES	40.	12.
Mercury	AA	0.5	0.6
Mercury	ES	<0.2	<0.2
Molybdenum	ES	220.	220.
Nickel	AA	50.	30.
Nickel	ES	88.	44.
Nitrogen	COU	3000.	3000.
Potassium	ES	6600.	4400.

AA - Atomic Absorption
ES - Emission Spectrochemical
NA - Neutron Activation
IE - Ion Electrode
COM - Combustion
COU - Coulometric
G - Gravimetric

Table 6 (Cont'd)

Minor and Trace Elements In
Coal Feed (Lower Kittanning)
and Product Coal (from EPA), ppm

Element	Method	Untreated	Treated
Samarium	ES	<10.	<10.
Selenium	ES	<10.	<10.
Silicon	ES	66000.	68000.
Silver	AA	0.98	0.97
Silver	ES	1.	1.
Sodium	ES	660.	400.
Strontium	ES	44.	44.
Sulfur	COM	33000.	14000.
Tellurium	ES	<10.	<10.
Thorium	ES	<10.	<10.
Tin	ES	2.	2.
Titanium	ES	440.	400.
Vanadium	NA	22.	28.
Vanadium	ES	44.	44.
Zinc	AA	32.	20.
Zinc	ES	40.	22.
Zirconium	ES	<4.	4.

AA - Atomic Absorption
ES - Emission Spectrochemical
NA - Neutron Activation
IE - Ion Electrode
COM - Combustion
COU - Coulometric

Table 7

Trace Element Analysis
Fox Mine
Lower Kittanning Seam,
Clarion County, Pennsylvania (From Ref. 9)

Element	Average Values, ppm		ppm Change	% Change
	Untreated	Treated		
Ag	<.1	1.1 ± 1.1	+1.1 ± 1.1	N.D.
As	23.5 ± 1.5	1.4 ± 1.2	-22.1 ± 1.9	-94 ± 5
B	16 ± 1.7	13 ± 0	-3 ± 1.7	19 ± 9
Be	2.0 ± 0.6	0.6 ± 0.2	-1.4 ± 0.6	-70 ± 14
Cd	<.5	<.5	Ind	Ind
Cr	94 ± 4.3	40 ± 4.5	-54 ± 6.2	-58 ± 5
Cu	25 ± 2.4	14 ± 0.6	-11 ± 2.5	-44 ± 6
F	93.5 ± 2.1	82 ± 0	-11.5 ± 2.1	-12 ± 2
Hg	0.07 ± 0.01	0.15 ± 0.02	+0.08 ± 0.02	Gain
Li	4 ± 0.1	15 ± 0.6	+11 ± 0.6	Gain
Mn	24 ± 2.5	9 ± 1.2	-15 ± 2.8	-63 ± 6
Ni	147 ± 5.1	11 ± 1.1	-136 ± 5.2	-93 ± 1
Pb	5 ± 2.9	12 ± 0.6	+7 ± 3	Gain
Sb	<1	<1	Ind	Ind
Se	17 ± 7.2	<2.5	-15 ± 7.2	-85%
Sn	<2.5	<2.5	Ind	Ind
V	94 ± 11.8	115 ± 10	+21 ± 15	Gain
Zn	105 ± 7.1	11 ± 0.6	-94 ± 7.1	-90 ± 1

8. PROCESS ALTERNATIVES AND IMPROVEMENTS

One process alternative is the use of a settling tank instead of hydroclones to reduce the water content of the solids from the reactor prior to filtration. This technique was suggested in reference 8. The use of a settling tank allows part of the reaction to take place in the tank and reduces the size of the reactors. A cost/efficiency analysis would be required to determine the best solids concentration technique.

Another process alternative discussed in reference 8 is the use of submerged combustion instead of steam to evaporate water before filtering the iron sulfates in the Iron Sulfate Recovery Section. Although a detailed cost comparison would be necessary to decide which method of evaporation is cheapest, it is felt that the use of steam would be the better of the two. Low pressure steam can be made available from the utilities area to give a steam balance and condensation of moisture from the large volume of combustion gas is then unnecessary.

It is felt that recovery of sulfur in a fractionation unit is preferable to the use of a pressure filter as suggested in Reference 8. Very little additional heat input is necessary for evaporation of the solvent. Again, however, a cost/efficiency evaluation would be necessary before a choice of units could be made.

If the Meyers process were located near a large power plant, steam and power requirements might be purchased. Since the plant location is unknown, the production of steam and power has been included in the present design.

As indicated previously, it will be necessary to prevent excessive buildup of solids dissolved from the coal. It is possible that the soluble materials can be allowed to buildup to the point where they begin to precipitate with the iron sulfates in the Iron Sulfate Removal Section. If this steady-state concentration does not affect the operability of the process, then the minor and trace elements can be disposed of with the iron sulfates. Otherwise, a purge stream from the process water circuit must be included. One simple method of accomplishing this is to replace the Iron Sulfate Recovery Section shown in figure 2 with the system shown in figure 3. The composition of the streams in figure 3 are given in Table 8.

In this disposal scheme, a fraction (0.3635) of stream 12, leach solution from filtration in the Sulfur Removal Section, sufficient to remove the necessary iron, is purged as stream B in figure 3. The remainder is passed to a reactor where sulfuric acid and oxygen are added to convert sufficient ferrous sulfate to ferric sulfate to give the iron compositions of stream 11 in figure 2. Sufficient water is flashed to give the water content of stream 11 and this stream is then returned to the Reaction Section. Make-up water (Stream G) is added to the overhead from the flash tank (stream F) to give stream 16 of figure 2 and this is passed to the Sulfur Recovery Section. Alternatively, stream A of figure 3 could go first to the Water Flash Tank and the bottoms could be returned to the Reaction Section. In this case, the sulfuric acid and extra oxygen would be added directly to the Reaction Section.

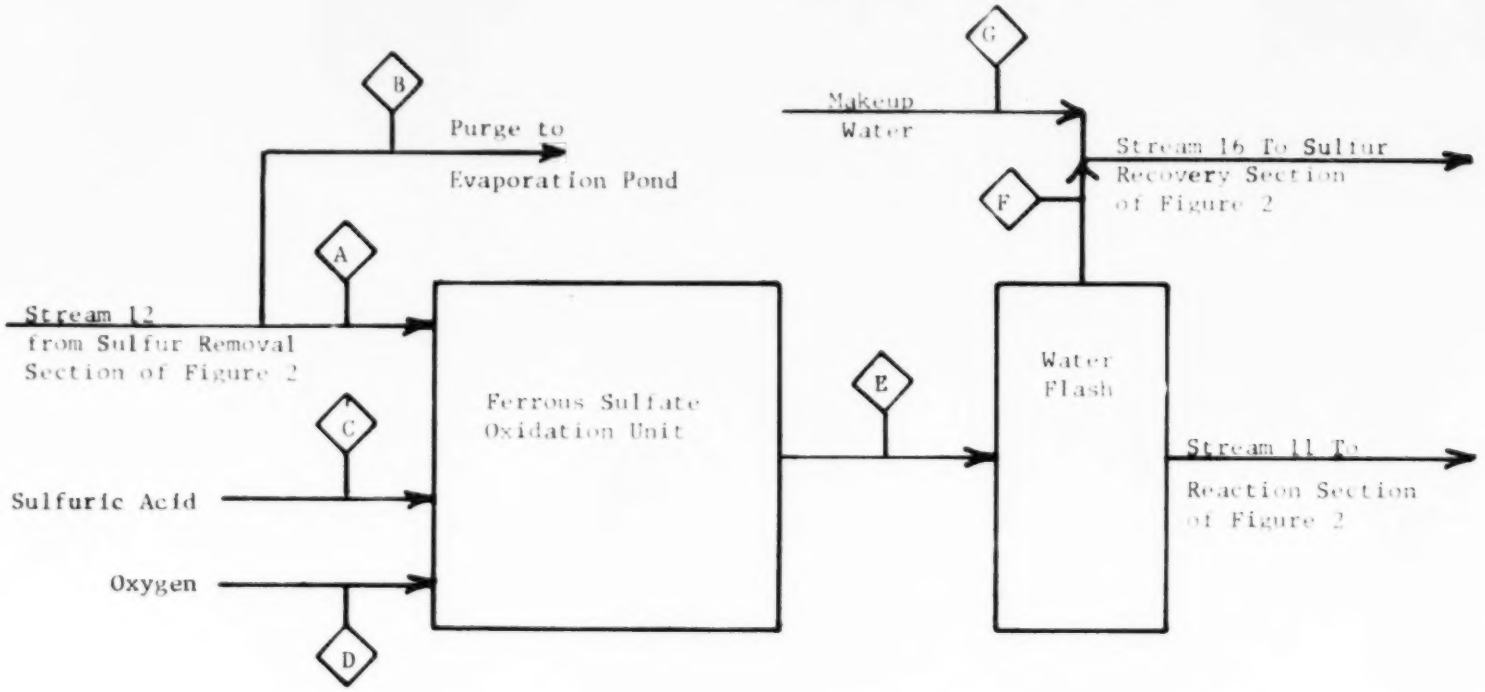

Figure 3

Flow Plan for Removal of Dissolved Solids and Iron from Meyers Process

Table 8

Material Balance for Removal of Dissolved Solids and Iron From Meyers Process, lb/hr

Stream → / Chemical ↓	12	A	B	C	D	E	F	G	11	16
$FeSO_4$	9,660	6,149	3,511	--	--	1,000	--	--	1,000	--
$Fe_2(SO_4)_3$	38,998	24,824	14,174	--	--	31,600	--	--	31,600	--
H_2SO_4	2,200	1,400	800	2,462	--	2,200	--	--	2,200	--
H_2O	231,000	147,038	83,962	--	--	147,343	20,743	83,657	126,600	104,400
O_2	--	--	--	--	271	--	--	--	--	--
TOTAL	281,858	179,411	102,447	2,462	271	182,143	20,743	83,657	161,400	104,400

The debits incurred by this scheme are:

- An increase in oxygen consumption of 271 lb/hr
- An additional 83,657 lb/hr. of process water
- Addition of 2462 lb/hr of H_2SO_4
- An evaporation pond for the purge

The credits for this scheme are:

- Savings of cooling water
- Steam savings for evaporation of approximately 31,000 lb/hr of water
- No filter required for iron removal

It is assumed that the purge stream will go to an evaporation pond for containment of the solids and acid. If neturalization of the acid is necessary, then limestone will be required.

The concentration of dissolved solids will be the weight of solids dissolved from the entering coal divided by the quantity of water purged in stream 2 of figure 3 (83,962 lb/hr of water). Thus, if 1% (ex pyrites) of the feed coal is dissolved, the concentration in stream 2 is 23,820 ppm. The concentration of dissolved solids will vary with the coal feed used. Thus, it has been found that the quantity of ash removed, other than that representing pyrites, varys over a ten-fold range (Ref. 8, Table 13). To increase the dissolved solids content of the recycle water stream, it would be necessary to remove part of the iron sulfate by precipitation and filtration as in the basic case. To decrease the solids content it would be necessary to purge a larger stream from the system. The iron content could be madeup by precipitation of part of the iron from the purge stream, filtering it and returning it to the system.

A different technique for removing dissolved solids has been suggested in a recent design for the Meyers process[12]. This involves evaporation of a portion of the sulfate stream to essentially dryness whereupon the dissolved solids are removed with the iron sulfate.

The process alternatives and improvements are summarized in Table 9.

Table 9

Process Alternatives and Improvements

- Use settling tank rather than hydroclones to increase solids content in stream from reactor.
- Use submerged combustion to evaporate water from iron sulfates solution.
- Use of a filter to remove elemental sulfur rather than fractionation.
- Purchase steam and electricity.
- Purge iron from system in solution form to remove other dissolved solids.
- Evaporate a portion of the iron sulfate to dryness and remove solids.

9. PROCESS DETAILS

Power and steam production and requirements are shown in Table 10. The plant water balance is given in Table 11.

Table 10

Power and Steam Balance

Utilities	Electricity kW	600 psia Steam lb/hr.	115 psia Steam lb/hr.
Consumed			
Coal Preparation	2,740		
Steam to vaporize H_2O in plant			120,000
Oxygen Plant		23,000	
Power generation		97,000	
Cooling water pumps	420		
Cooling tower fans	260		
Boiler feed water pumps	110		
Rest of plant	1,000		
	4,530	120,000	120,000
Produced			
Power generation	4,530		
Steam plant		120,000	
From 600 psia Steam			120,000
	4,530	120,000	120,000

Table 11

Water Balance*

		lb/hr
Into Plant		
To makeup water treatment		153,850
In feed coal		20,000
In air to O_2 plant		170
	TOTAL IN	174,020
Out of Plant		
Cooling tower		135,560
Evaporation	100,000	
Drift	14,160	
Blow Down	21,400	
In total product coal		37,600
Vents		860
Iron sulfate product		(Unknown)**
	TOTAL OUT	174,020

* Excluding potable water and sanitary sewer.

** Any positive value would necessitate increased makeup.

10. TECHNOLOGY NEEDS

In order to more clearly define and quantify potential pollution problems, certain areas will have to be investigated in more detail than has been done heretofore. The major items requiring further investigation are discussed here.

One major item that may need further work involves overall sulfur removal from coal. As indicated previously, some coals can be treated to meet existing regulations (Refs. 10 and 11). However, in the work reported in Refs. 8 and 9, the total sulfur content of the treated coal is still about 1% or more. Although in most cases this represents mainly organic and sulfate sulfur, the treated coals could not meet the present Federal standard of 1.2 lb SO_2/MM Btu. For those coals, if this process were used to remove pyritic sulfur, an organic sulfur removal step would also be needed.

Another area for clarification, for the particular coal of interest, is the loss of ferric ion due to reactions with the coal matrix. The experimental reduction of ferric ion over theoretical has been shown to range from 20% excess for a Lower Kittanning coal to 140% for a Herrin No. 6 sample. These losses were not included in the present design. The effect of this loss would be an increase in oxygen consumption for the process together with a resulting increase in steam requirements as well as consumption of sulfuric acid. The increase in high pressure steam requirements would affect the balance in high and low pressure steam and this would have a major affect on the overall process design.

The concentration of dissolved solids in the process water streams should be more clearly defined. This concentration will have an effect on the quantity of purge from the system and hence on the quantity of makeup water and chemicals.

The concentration of organic solvent in the effluent purge water should be investigated. Naphtha has a measurable solubility in water and can appear in the purge stream. It may be necessary to remove this organic material by stripping or biological oxidation prior to removing the purge stream to an evaporation pond.

Another area that requires investigation is the buildup of organic impurities in the recycled organic solvent. Since the coal matrix is affected by the oxidizing action of ferric sulfate, it is possible that soluble organic materials may be extracted along with the sulfur. Such material could be removed as a contaminant in the sulfur or could build up in the naphtha. The former would represent a product quality problem and the latter would require a solvent purge with a purification step added.

Filter size and wash water requirements need to be defined more carefully. The possibility of iron oxide formation which could affect filtration rates has been pointed out (Ref. 8, p. 71). The wash water requirement for laboratory experiments is much larger than that assumed in the present design. A large increase in wash water requirement would necessitate major design changes.

The quantity of water in the product coal requires quantification. The suitability of the product for furnace fuel could be affected onsiderably if the product is sticky. If the moisture in the product is too great, drying may have to be provided and this would decrease the thermal efficiency.

A better method of disposal of the iron sulfates is needed since the quantity of this byproduct for the present case is almost 200 tons per day. Although a holding pond has been assumed in this study for iron sulfate disposal, it would be preferable if a method were available to recover the iron and sulfur values from the material. In any case, the quantity of surface moisture and water of hydration leaving with the iron sulfates should be defined as this will affect the quantity of make up water.

11. QUALIFICATIONS

As pointed out, this study does not consider cost or economics. Also, areas such as coal mining and general offsites are excluded. These will be similar and common to all conversion operations.

The study is based on a specific process design and coal type, with modifications as discussed. Plant location is an important item of the basis and is not always specified in detail. It will affect items such as the air and water conditions available, and the type of pollution control needed. For example, this study is based on high sulfur eastern coal, although it can be used on low sulfur western coal. Because of variations in such basis items, great caution is needed in making comparisons between coal conversion processes since they are not on a completely comparable basis.

Some other conversion processes are intended to make SNG or low-Btu gas fuel, and may make appreciable amounts of by-products, such as tar, naphtha, phenols, and ammonia. Such variability further increases the difficulty of making meaningful comparisons between processes.

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* The section numbers of references 1, 2, 3, and 4 have been changed to 1 through 4, respectively.